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[Title of Invention]            Scintillator and Radiation Detector,  
and Radiation Inspecting Device

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[Document Name] claims

[Claim 1] A scintillator consisting of a crystal of  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$  ( $0 < x < 0.5$ ).

[Claim 2] The scintillator according to claim 1 characterized in that  $0.03 < x < 0.2$ .

[Claim 3] The scintillator according to claim 1 or 2 characterized in that said crystal is grown by the micro pulling down method, Czochralski method, the floating zone method, or Bridgman method.

[Claim 4] A radiation detector consisting of a combination of the scintillator according to any one of claims 1 to 3 and a light responding means.

[Claim 5] A radiation inspecting device having the radiation detector according to claim 4 as the radiation detector.

[Claim 6] The radiation inspecting device according to claim 5 characterized in that said radiation inspecting device is an X-ray CT scanner.

[Claim 7] The radiation inspecting device according to claim 6 characterized in that said radiation inspecting device is PET (positron emission tomography).

[Document Name] specification

[TITLE OF THE INVENTION]

Scintillator and Radiation Detector, and Radiation  
Inspecting Device

[FIELD OF THE INVENTION]

[0001]

This invention relates to various radiation detectors,  
scintillators and radiation detectors, and radiation  
inspecting devices.

[BACKGROUND OF THE INVENTION]

[0002]

Patent document 1: Japanese Patent Laid-Open No.5-87934

The scintillator crystals are used in many fields as  
detectors of various radiations, such as X-ray and  $\gamma$ -ray.  
Required characteristics for the scintillator crystals are  
somewhat changed depending on their uses, but generally  
involve :

a high density;

a large fluorescence output by applying radiation;

a high fluorescence attenuation rate:

good radiation tolerance:

a crystal having no deliquescence or cleavability, and  
easy to be processed

In these days, taking into account these requirements,  
crystals adopting active Ce with high attenuation rate (20

- 60 ns) are often used. For example,  $\text{Gd}_2\text{SiO}_5:\text{Ce}$  (GSO),  $\text{Lu}_2\text{SiO}_5:\text{Ce}$  (LSO) and the like are used in the medical diagnostic equipments, such as PET (positron emission tomography), but the above-mentioned required characteristics are not necessarily satisfied sufficiently and there are problems that: in GSO, the crystal raising requires high techniques due to strong crystal anisotropy and becomes the hindrance of cost reduction, and in LSO, the fluorescence output has variations between samples.

[0003]

Also, as the scintillators using Pr, Ce and F, those consisting of  $\text{Gd}_2\text{O}_2:\text{Pr}$ , Ce and F have been known as described in Patent document 1.

[0004]

However, even among the scintillators according to Patent-documents 1, scintillators with stable characteristics (especially fluorescence output) have not yet been obtained.

[DISCLOSURE OF THE INVENTION]

[PROBLEMS TO BE SOLVED BY THE INVENTION]

[0005]

The present invention aims to provide a scintillator having high and stable fluorescence output, crystal raising thereof being comparatively easy.

[0006]

The present invention aims to provide a radiation detector having high and stable detection sensitivity.

[0007]

The present invention aims to provide a medical radiodiagnosis device which can obtain captured images with high-resolution.

[MEANS FOR SLOVING THE PROBLEMS]

[0008]

The scintillator of the present invention is characterized by consisting of a crystal of  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$  ( $0 < x < 0.5$ ).

[0009]

In particular,  $0.03 < x < 0.2$  is preferred.

[0010]

The radiation detector of the present invention is characterized by consisting of a combination of the above-mentioned scintillator and a light responding means.

[0011]

The radiation inspecting device of the present invention is characterized by being provided with the above radiation detector as its radiation detector.

[EFFECT OF THE INVENTION]

[0012]

According to the present invention, it has become possible to provide a scintillator wherein the performance is high with respect to luminescence intensity and attenuation rate.

[0013]

It has become possible to provide a radiation detector

whose detection sensitivity is high and fluorescence output is stable.

[0014]

It has become possible to provide a radiation inspecting device which can obtain captured images with high-resolution. As the radiation inspecting device, PET (positron emission tomography) is preferred.

[THE BEST MODE FOR CARRYING OUT THE INVENTION]

[0015]

(Scintillator composition :  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$ )

The scintillator emits a light in ultraviolet or visible region, when it is irradiated with a light or a radiation.

[0016]

The scintillator of the present invention consists of a crystal having composition of  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$ , wherein  $0 < x < 0.5$ .

[0017]

When cerium is not doped to praseodymium fluoride (in the case of  $x = 0$ ), if X-ray is irradiated thereon, luminescence originating from  $\text{Pr}^{3+}$  is observed at 400 nm, and the attenuation time thereof is very slow, about 600 ns. However, when cerium is doped, luminescence at 400 nm originating from  $\text{Pr}^{3+}$  decreases, and instead, luminescence originating from  $\text{Ce}^{3+}$  appears near 290 nm. The attenuation time thereof is from 17 to 17.5 ns, which is faster than 27 ns of  $\text{CeF}_3$  used for energy measurement of high energy  $\gamma$ -ray.



As the added cerium concentration is increased, the luminescence intensity at 290nm further increases and the luminescence at 400 nm loses its intensity.

[0018]

Particularly, in the case of  $0.03 < x < 0.2$ , the luminescence intensity at 290 nm becomes large to give the luminescence intensity equivalent to or beyond that of  $\text{CeF}_3$ , which is comparatively strong among the fluorides. However, when  $x$  is 0.5 or larger, the luminescence intensity at 290 nm decreases.

[0019]

(Manufacturing method of the scintillator)

The scintillator crystal of the above-mentioned composition is preferred, but not limited, to be grown by the micro pulling down method or by the general crystal growing methods such as the Czochralski method, the Bridgman method, or the floating zone method.

Among them, the micro pulling down method allows crystal to grow at a speed one or two digit higher than usual melt growth method. Therefore, the required time to grow crystal is short, and single crystals of significant size and quality can be obtained from small amount of materials.

[0020]

For example,  $x$  can be controlled by raw material. What is necessary is just to calculate the amounts of the raw materials,  $\text{PrF}_3$  and  $\text{CeF}_3$ , so that predetermined  $x$  may be obtained. Predetermined amounts of  $\text{PrF}_3$  and  $\text{CeF}_3$  may be

mixed and fused in a crucible.

Since the conventional scintillator was mainly manufactured by sintering, the composition control thereof has been difficult, but by manufacturing the scintillator by the above-mentioned crystal growing method, production of the crystal whose compositions are fully controlled can be attained.

[0021]

(Radiation detector)

The radiation detector of the present invention consists of a combination of the scintillator and a light responding means.

The light responding means converts luminescence from the scintillator into an electric signal. For example, photoelectric conversion elements, such as a photodiode may be used. A photomultiplier element may be also provided.

[0022]

(Radiation Inspecting device)

Providing a radiation detector as a radiation detector will make an apparatus effective in detecting the radiations in various fields.

A method for acquiring structural or compositional information of a subject as a two-dimensional image by irradiating the subject with various radiations such as X-ray, neutron beam and  $\gamma$ -ray, and measuring the intensity distributions of the radiation penetrated through the subject with a radiation detector (radiography), can be

widely utilized, as X-ray diagnostic devices in the medical field, dangerous materials detecting devices for baggage, and nondestructive inspection devices for various constructions.

[0023]

Also, the neutron radiography is a method for acquiring structural or compositional information of the subject as two-dimensional images, by detecting the intensity distribution of the thermal neutron beam which is attenuated by penetrating through the subject. It is effective in examination of hydrogen containing compounds and composite materials comprising metals and the light element substances which are difficult to be examined by the X-ray or the  $\gamma$ -ray, and is utilized as an effective examination method in the wide fields such as plant apparatus, airplanes, auto parts and the like.

[0024]

An X-ray diagnostic device (CT scanner) arranges many X-ray detectors around the patient as a subject, the signals of the penetrated X-rays received by these detectors are reconstructed as tomograms by computer-operated processing, and the tomograms are displayed on an image display apparatus such as CRT or obtained as a photograph. Since the tomograms by this X-ray diagnostic device are obtained as slice images of a human body unlike the usual X-ray photographs and the like, it becomes possible to diagnose the disease in the depths of the human body such as

internal organs with high precision.

[0025]

Also the radiation detector of the present invention is applicable in the environmental measurement devices and in the fields of various computer processing radiography which detect nuclear radiations.

[0026]

(COMPARATIVE EXAMPLE 1)

Among the crystals of the present invention,  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$ , that of  $x=0.01$  was grown by the fluoride micro PD method. High purity  $\text{PrF}_3$  and  $\text{CeF}_3$  as the materials were weighed and mixed, and then were charged in a high purity platinum crucible with a small pore at the bottom. As shown in Fig. 1, a seed, a stage, after heater, thermal insulating material, and the crucible charged with materials were arranged, and were heated to  $700^\circ\text{C}$  in vacuo exhausted to about  $1 \times 10^{-3}$  Pa with an oil rotary pump and an oil diffusion pump. Then, the inside of the chamber was replaced by Ar gas. The sample was then melted by being heated to about  $1450^\circ\text{C}$  with a high frequency coil. The bottom of the crucible was monitored with a CCD camera. When the melt appeared from the small pore at the crucible bottom, a seed crystal was attached to it and was pulled down at a rate  $0.05\text{--}0.5$  mm/min to be solidified. As a result, a green clear crystal with  $\phi 3$  mm and 50mm in length was obtained. When the obtained crystal was irradiated by X-ray at room temperature, strong luminescence was observed at 290 nm,

and also at 400nm.

[EXAMPLE 1]

[0027]

Among the crystals of the present invention,  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$ , that of  $x=0.03$  was grown by the fluoride micro PD method. The crystal was grown in the same way as Comparative Example 1, and a green clear crystal 50 mm in length was obtained. When the obtained crystal was irradiated by the X-ray at room temperature, strong luminescence was observed at 290 nm, which was stronger than in Example 1. Luminescence was also observed at 400nm, but it was smaller than in Example 1. In this way, the effect of increase in the concentration of added cerium was observed. When an attenuation time of the luminescence at 290 nm was measured by ultraviolet light excitation, it was 17 - 17.5 ns. It was 20.5 ns, when the attenuation time by X-ray excitation was measured.

[EXAMPLE 2]

[0028]

In this example,  $x$  was further changed among 0, 0.001, 0.01, 0.03, 0.06, 0.1, and 0.2. Crystal was grown in the same way as Comparative Example 1, and a green clear crystals of 20 - 50 mm in length were obtained. The above luminescence data were shown in Table 1.

Table 1.

X (in $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$ )	Luminescence intensity at 290 nm (originated from $\text{Ce}^{3+}$ ) * <sup>1</sup>	Luminescence intensity at 400 nm (originated from $\text{Pr}^{3+}$ ) * <sup>2</sup>
0	0	100
0.001	10	90
0.01	50	30
0.03	70	15
0.06	100	0
0.1	120	0
0.2	100	0

\*1. Setting  $\text{CeF}_3$  as 100

\*2. Setting  $\text{PrF}_3$  as 100.

## [EXAMPLE 3]

[0029]

In this example, the crystal of  $x = 0.1$  in  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$ , was grown by the Czochralski method. Materials, high purity  $\text{PrF}_3$  and  $\text{CeF}_3$ , were weighed and mixed, and then were charged in a carbon crucible. This was installed in a growing furnace, heated to 700 °C in vacuo exhausted to about  $1 \times 10^{-3}$  Pa with an oil rotary pump and an oil diffusion pump. Then, the inside of the chamber was substituted by Ar gas, and heated to about 1450 °C with a high frequency coil to melt the sample. When the temperature became stable, the sample was contacted with the seed crystal, and the crystal was grown at the pull-up speed of 1 mm/h, rotating at 10-20 rpm. A green clear crystal without crack with 50 mm in

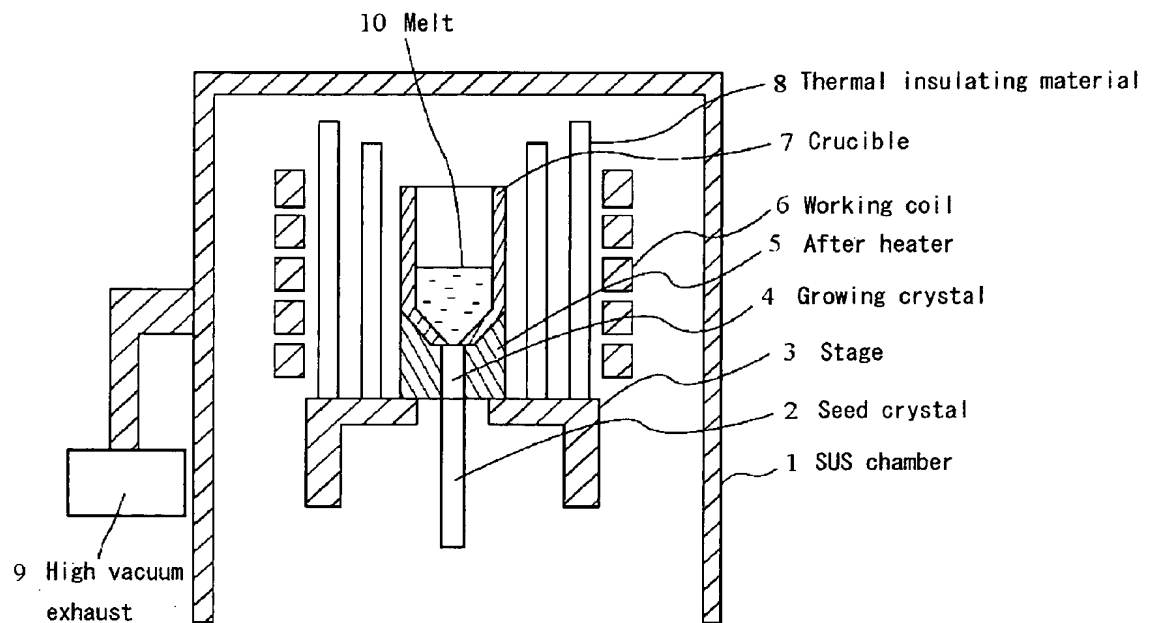
diameter and about 150 mm in length was obtained. When the obtained crystal was irradiated by the X-ray at room temperature, strong luminescence was observed at 290 nm, and a similar result to Example 2 was obtained.

[BRIEF EXPLANATION OF THE DRAWINGS]

[0030]

Fig. 1. A schematic diagram of an atmosphere control high-frequency-induction-heating type micro pulling down apparatus.

FIG.1





[Document name] Abstract

[Abstract]

[Problem] A material for a scintillator with high performance with respect to luminescence intensity and attenuation rate, whose crystal growing is comparatively easy, is provided.

[Means of Solution] The scintillator is a crystal consisting of  $\text{Pr}_{1-x}\text{Ce}_x\text{F}_3$  ( $0 < x < 0.5$ ), and emits light in ultraviolet and visible regions by lights or radiations.

[Chosen Drawing] FIG. 1